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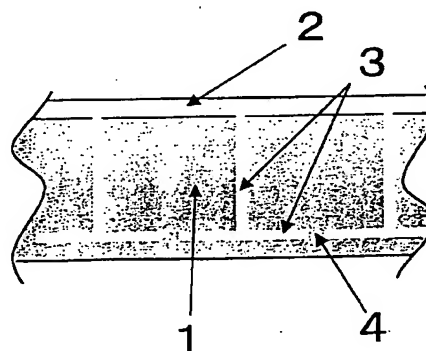
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**(54) POLYPROPYLENE FILM AND CAPACITOR MADE BY USING THE SAME AS THE DIELECTRIC**

(57) To provide a biaxially oriented polypropylene film excellent in heat resistance and long-term dielectric properties at high temperatures, less in insulation defects, and excellent in the impregnation of an insulating oil into the clearance between film layers and swelling resistance when immersed in the insulating oil, and also to provide a capacitor excellent in heat resistance, dielectric properties, corona resistance, long-term thermal durability and electric current resistance, using the polypropylene film as dielectric.

The film of the present invention is a biaxially oriented polypropylene film characterized by being 98 to 99.5% in isotacticity, 99% or more in isotactic pentad fraction, 30 ppm or less in ash content and 0.01 to 0.4  $\mu\text{m}$  in the center line average surface roughnesses on both sides.

FIG. 2



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## Description

## Technical Field

5 The present invention relates to a biaxially oriented polypropylene film and a capacitor using it as a dielectric, particularly a biaxially oriented polypropylene film excellent in heat resistance and dielectric properties, less in insulation defects and excellent in the impregnation of an insulating oil into the clearances between film layers and swelling resistance when immersed in the insulating oil, and a capacitor excellent in dielectric properties, corona resistance, long-term thermal durability and electric current resistance, using the film as dielectric.

## Background Arts

10 Biaxially oriented polypropylene films are widely used for packaging, tapes and capacitors, since they are excellent in optical properties such as clarity and gloss, mechanical properties such as tensile strength and elongation, moisture barrier properties and electrical properties.

15 Biaxially oriented polypropylene films are one of major materials used as dielectric of film capacitors, but since they are lower in heat resistance compared to polyester films, another major material, the capacitors obtained by using them are limited to about 85°C in the maximum operating temperature. As for the cause, if the operating temperature becomes high, the dielectric strength peculiar to the polypropylene films drastically declines due to the influence of the amorphous domains of the film and impurities, particularly making them unable to withstand long-term use in some cases.

20 On the other hand, as electric apparatuses are being downsized, devices are assembled at higher densities and exposed to higher temperatures, and there is a strong demand to use the conventional polypropylene film capacitors at higher operating temperatures. Especially for capacitors to be mounted on AC circuits, polypropylene films with a feature of being small in dielectric loss have been used as dielectric for the necessity of inhibiting the heat generation from inside the capacitor devices, but since the temperature of the environments surrounding AC circuits becomes higher, it is now difficult to use the polypropylene films for such an application. This is one of the factors to inhibit the downsizing of electric apparatuses equipped with polypropylene film capacitors. As for the reason, even though a circuit has been located with a certain distance kept away from the heat source or insulated from the heat source by a heat insulator lest the circuit should be heated by the heat source, there is an increasing demand to install such a circuit very close a heat source without using any heat insulators for meeting the demand for downsizing. This demand is typically seen for the AC circuits used in the illumination stabilizers of street lighting, etc. and the control circuits of motors. To meet this demand, it is necessary that the polypropylene film maintains its performance even at temperatures higher than 85°C, the maximum operating temperature of conventional polypropylene film capacitors, for a longer period of time.

25 So, the polypropylene film used as a dielectric is demanded to be (1) moderately small in mechanical deformation, i.e., heat shrinkage when rapidly heated within a short time, (2) excellent in electrical properties at a high temperatures and (3) small in the decline of electrical properties at high temperatures with the lapse of time.

30 As for the reason for (1), when a capacitor device is produced, the polypropylene film wound with an electrode is generally annealed at a certain temperature, to be moderately thermally shrunken for tightening the winding to maintain its shape and to squeeze out the air between film layers, and in this case if the heat shrinkage is too large, the device may be deformed to lower the capacity of the capacitor or may be destroyed. If the heat shrinkage is too small, the tightening of the winding may be insufficient or the dielectric loss may increase to destroy the device when used at a high temperature for a long time.

35 It is sometimes practiced to impregnate the capacity device with an insulating oil, for inhibiting the corona otherwise generated, the capacity decline and dielectric breakdown of the capacitor in continuous use and the increase of leak current. In such impregnation with an insulating oil, (4) it is necessary to design the film surface and (5) it is necessary to inhibit the dimensional change and swelling caused by the insulating oil, in order to ensure that the insulating oil is uniformly impregnated into the clearances between film layers when the polypropylene film used as a dielectric is wound as a capacitor device.

40 Furthermore, while the improvement of above mentioned properties is desired to allow the downsizing of electric apparatuses as described above, there is also a more intensive demand for further downsizing the capacitor itself. For this purpose, the electrostatic capacity of the capacitor per unit volume must be increased, and the film as the dielectric must be thinned. This requires the film to have a higher dielectric strength even at room temperature for meeting the above mentioned demand for a thinner film, though the film has been conventionally designed to be thicker to provide an allowance in dielectric properties.

45 Especially the capacitor for a high frequency circuit using a polypropylene film as dielectric is more demanded to be higher in dielectric properties and electric current resistance.

To solve the problem, Japanese Patent Laid-Open (Kokai) No. 6-236709 discloses a high molecular insulating

material low in ash content, of 1 to 10 wt% in boiling n-heptane soluble content, hence excellent in processability, and excellent in dielectric properties in a temperature range from room temperature to 80°C, and it is stated that a material of 90% or more in the isotactic pentad fraction of the boiling n-heptane insoluble portion is preferable.

Japanese Patent Laid-Open (Kokai) No. 7-25946 proposes a propylene polymer of 80 wt% or more, especially preferably 96 wt% or more in boiling heptane insoluble content and 0.970 to 0.995 in the isotactic pentad fraction of the boiling heptane insoluble component, and also proposes a molding obtained by using the polymer.

However, biaxially oriented polypropylene films simply high in the isotactic pentad fraction of the boiling n-heptane insoluble portion as proposed here are insufficient in the dielectric properties at temperatures higher than 85°C intended in the present invention, and also in the long-term heat resistance of the capacitor device using the film as dielectric. That is, though the biaxially oriented polypropylene films high in stereoregularity obtained by said conventional techniques are anyway high in the isotactic pentad fraction of the boiling n-heptane insoluble portion, the isotactic pentad fraction as a film is destined to be low since the isotactic pentad fraction of the n-heptane soluble portion is low. As a result, the stereoregularity is insufficient. In addition, a so-called high crystalline biaxially oriented polypropylene film very high in isotacticity is very poor in film formability since it is insufficient in stereoregularity, and no industrially useful technique has been established to produce a biaxially oriented polypropylene film excellent in heat resistance and dielectric properties.

As a technique to overcome this disadvantage, Japanese Patent Publication (Kokoku) No. 4-28727 proposes a crystalline polypropylene film excellent in moldability by keeping the isotactic pentad fraction in a range of 0.960 to 0.990 and keeping the total amount of the extracts obtained by sequentially extracting with boiling n-hexane and boiling n-heptane in a range of 3.0 to 6.0%. However, the film is insufficient in the isotactic pentad fraction as well as in the dielectric properties at high temperatures.

Furthermore, Japanese Patent Laid-Open (Kokai) No. 5-217799 proposes a metallized film capacitor using a high-modulus metallized film obtained by vapor-depositing a metal on a high-modulus polypropylene film with a specific thermal deformation temperature and a specific Young's modulus and high in crystallinity and good in stereoregularity. However, the stereoregularity is only about 90% at the highest and the dielectric properties at high temperatures is insufficient.

Moreover, Japanese Patent Laid-Open (Kokai) No. 7-50224 proposes a metallized polypropylene film of 4.0% or less in the heat shrinkage at 120°C in the machine direction and 0.8% or less in the transverse direction. However, the film is at the conventional levels in isotacticity and stereoregularity, and cannot be said to be sufficient in the dielectric properties at high temperature intended in the present invention to meet the advanced demand in future.

#### Disclosure of the Invention

The inventors found that if the polypropylene film is highly controlled in isotacticity and stereoregularity, a polypropylene film with very high isotacticity can be formed, that if proper film forming conditions are adopted, the dielectric properties peculiar to a polypropylene film can be further improved though this cannot be achieved by the conventional techniques, and furthermore that the capacitor obtained using the film is restrained in the decline of properties at high temperatures and also in the long-term deterioration at high temperatures and is small in the dispersion of quality. Thus, the present invention has been completed.

An object of the present invention is to provide a polypropylene film excellent in heat resistance and the long-term dielectric properties at high temperatures and room temperature, less in insulation defects, and excellent in the impregnation of insulating oil into the clearance between film layers and swelling resistance when immersed in the insulating oil.

Another object of the present invention is to provide a capacitor excellent in heat resistance, long-term dielectric properties at high temperatures and room temperature, corona resistance and electric current resistance, using said polypropylene film as dielectric.

The above objects of the present invention can be achieved by a biaxially oriented polypropylene film, characterized by being 98 to 99.5% in isotacticity, 99% or more in isotactic pentad fraction, 30 ppm or less in ash content, and 0.01 to 0.4  $\mu\text{m}$  in the center line average surface roughnesses on both sides of the film. The capacitor using the polypropylene film of the present invention as its dielectric can be suitably used as a capacitor for a heat resistant AC circuit, a capacitor for a high frequency circuit or a capacitor for a heat resistant DC circuit.

#### Brief Description of the Drawings

Fig. 1 is a plan view showing a polypropylene film with a metallic layer on one side as an embodiment of the present invention.

Fig. 2 is a plan view showing a polypropylene film with a metallic layer on one side and with the electrode separated like isles in the machine direction of the film as another embodiment of the present invention.

Fig. 3 is a plan view showing a polypropylene film with a metallic layer on one side and with the electrode having a plurality of safety function portions in the transverse direction of the film.

In the drawings, symbol 1 denotes a metallic layer (internal electrode); 2, an insulation groove (margin); 3, insulation grooves (to separate the internal electrode like isles); and 4, bottle necks (safety function portions), respectively.

#### The Most Preferred Embodiments of the Invention

The polypropylene used for the polypropylene film of the present invention is mainly propylene homopolymer, but as far as the objects of the present invention are not impaired, a comonomer such as another unsaturated hydrocarbon, etc. can also be copolymerized, or another copolymer of propylene can also be blended.

The monomers which constitute the copolymers and blends include, for example, ethylene, propylene (in the case of a blend with another copolymer), 1-butene, 1-pentene, 3-methylpentene-1, 3-methylbutene-1, 1-hexene, 4-methylpentene-1, 5-ethylhexene-1, 1-octene, 1-decene, 1-dodecene, vinylcyclohexene, styrene, allylbenzene, cyclopentene, norbornene, 5-methyl-2-norbornene, etc. As for the amount, in view of dielectric properties and heat resistance, the amount of the comonomer to be copolymerized is preferably less than 1 mol%, and the amount of another copolymer to be blended is preferably less than 10 wt%.

In the present invention, the isotacticity of the polypropylene film must be 99.5% or less in view of film formability.

The isotacticity in this case is defined as the weight percent of the insoluble portion of the film after extraction in boiling n-heptane based on the weight of the film before extraction. If the isotacticity is too high, stretchability is so low as to make film formation very difficult when a biaxially oriented film is produced, as described in Japanese Patent Laid-Open (Kokai) No. 6-236709. Furthermore, in view of heat resistance and dielectric properties, the isotacticity must be 98% or more.

For good film formability, heat resistance and dielectric properties, the isotacticity is preferably 98.5 to 99.5%, more preferably 98.7 to 99.3%.

To obtain a polypropylene film with such isotacticity, for example, the polypropylene resin selected as the raw material should be low in the content of low molecular weight components and low in stereoregularity, that is, moderately low in the rate of the so-called atactic portion likely to be dissolved in boiling n-heptane.

In the present invention, the stereoregularity of a polypropylene film can be evaluated in terms of the pentad fraction determined from the methyl absorption peak measured by  $^{13}\text{C}$ -NMR. In general, the conformations of the five repeating units (pentad) in a polypropylene molecular chain include mmmm, mmmr, rmmmr, ..., rrrr, mrrr, mrrm, where m denotes the meso conformation and r, the rasemo conformation. To obtain the pentad fraction of a polypropylene film, the proportions of the segments with the above respective conformations can be determined from  $^{13}\text{C}$ -NMR, as reported, for example, by T. Hayashi et al. [Polymer, 29, 138-143(1988)]. Of these proportions, the fraction of the conformation of mmmm to the absorption intensity of all the methyl groups, i.e., the isotactic pentad fraction (hereinafter may be simply expressed as mmmm) can be defined as the sum of three heptad fractions of m(mmmm)m, m(mmmm)r and r(mmmm)r.

The isotactic pentad fraction mmmm of the polypropylene film of the present invention is 99% or more. Since such a film is made of polypropylene constituted by molecules with very long isotactic segments, it can provide a film with high crystallinity, high heat resistance and excellent dielectric properties. The mmmm of the polypropylene film of the present invention is preferably 99.1% or more, more preferably 99.2% or more, further more preferably 99.3% or more in view of high heat resistance and excellent dielectric properties. To secure such stereoregularity, it is effective to highly control the stereoregularity of the polypropylene resin used as the raw material. Such a raw material can be produced by selecting the catalyst system (a solid catalyst and an external electron donor compound) and controlling the purities of the components of the catalyst system. If the polypropylene resin used as the raw material is higher in mmmm, the polypropylene film obtained tends to be higher in mmmm, but since the extreme thermal deterioration in the extrusion system of the raw material lowers mmmm, the structural design and extrusion conditions should be selected, for example, to avoid the long-time stagnation of the raw material in the high temperature extrusion system.

In the polymerisation process of the polypropylene used for the polypropylene film of the present invention, it is generally practiced to use a metal-containing compound as a catalyst and as required, to remove the residue after completion of polymerization. The residue can be evaluated by obtaining the amount of the metal oxide remaining after perfectly burning the resin. This called ash content.

The ash content of the polypropylene film of the present invention must be 30 ppm or less, preferably 25 ppm or less, more preferably 20 ppm or less. If the ash content is more than 30 ppm, the dielectric properties of the film declines, and the dielectric properties of the capacitor obtained by using the film also declines. To keep the ash content in this range, it is important to use a catalyst low in the residue, but it also can be adopted to decrease the contamination from the extrusion system at the time of film formation as far as possible, for example, by bleeding for more than 1 hour.

The polypropylene used for the polypropylene film of the present invention can contain publicly known additives, for example, a nucleating agent, antioxidant, thermal stabilizer, slip agent, antistatic agent, anti-blocking agent, filler, vis-

cosity improver, anti-coloring agent, etc. as far as the properties of the present invention are not impaired.

Among these additives, the selection of the antioxidant used and its amount are important for long-term heat resistance. The antioxidant added to the polypropylene film of the present invention is phenolic ones with large steric hindrance, and at least one of antioxidants used is preferably a high polymer of 500 or more in molecular weight to minimize the volatile loss at the time of melt extrusion.

Various antioxidants are available, and it is preferable, for example, to use 2,6-di-t-butyl-p-cresol (BHT; 220.4 in molecular weight) together with 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene (e.g., Irganox 1330 produced by Ciba Geigy; 775.2 in molecular weight) or tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane (e.g., Irganox 1010 produced by Ciba Geigy; 1177.7 in molecular weight), etc. The total content of the antioxidants is preferably 0.03 to 1.0 wt% (300 to 10000 ppm) based on the total amount of polypropylene. If less than 0.03 wt%, the long-term heat resistance may be poor, and if more than 1.0 wt%, these antioxidants may bleed out, to cause blocking at high temperatures, adversely affecting the capacitor device. The antioxidant content is more preferably 0.1 to 0.9 wt%, further more preferably 0.2 to 0.8 wt%.

As for antioxidants, it is preferable, in view of the long-term durability of the capacitor using the polypropylene film as its dielectric, that 2,6-di-t-butyl-p-cresol (BHT; 220.4 in molecular weight) showing better electrical properties is used together with 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene (e.g., Irganox 1330 produced by Ciba Geigy; 775.2 in molecular weight) and/or tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane (e.g., Irganox 1010 produced by Ciba Geigy; 1177.7 in molecular weight) by 0.05 to 0.35 wt% in total.

The addition of a nucleating agent affects the surface roughness and clarity of the film, but tends to worsen the dielectric strength. So, its content is preferably less than 0.1 wt%, and more preferably it is not substantially contained.

In the present invention, the intrinsic viscosity of the polypropylene excellent in stereoregularity used for the polypropylene film is not especially limited, but is preferably 1 to 10 dl/g in view of film formability. Furthermore, the melt flow rate at a load of 2.16 kg at 230°C is preferably 2 to 5 g/10 min in view of film formability. The intrinsic viscosity and the melt flow rate can be kept in the above ranges, for example, by controlling the average molecular weight and the molecular weight distribution.

The polypropylene film of the present invention can be obtained by biaxially orienting the raw material destined to have the above mentioned properties. If the film is not oriented, the film with high crystallinity, high heat resistance and high dielectric properties as intended in the present invention cannot be obtained. The biaxial orienting method can be any of simultaneous biaxial drawing by inflation, simultaneous biaxial drawing by a stenter or sequential biaxial drawing by stenters. Among them, in view of film forming stability, uniform thickness and the control of the surface roughnesses described later, sequential biaxial drawing by stenters can be preferably used for film formation.

In the present invention, the center line average surface roughnesses on both sides of the biaxially oriented polypropylene must be 0.01 to 0.4  $\mu\text{m}$ . If the center line average surface roughnesses are too large, air remains clearance between stacked film layers, to deteriorate the capacitor device, and when a metallic layer is formed on the film, the metal layer is damaged, to lower the dielectric strength and the device life at high temperatures, and to cause insulation defects when a voltage is applied. If too small on the contrary, the film is insufficiently slippery, to inconvenience handling, and when the capacitor device is impregnated with an insulating oil, the insulating oil does not uniformly impregnate into the clearance between the film layers, to greatly change the capacity in continuous use. The center line average surface roughnesses on both sides of the film are more preferably 0.03 to 0.3  $\mu\text{m}$ , further more preferably 0.04 to 0.25  $\mu\text{m}$ .

The biaxially oriented polypropylene film of the present invention is preferably 0.01 to 0.5  $\mu\text{m}$ , more preferably 0.02 to 0.4  $\mu\text{m}$ , most preferably 0.03 to 0.3  $\mu\text{m}$  in the difference ( $\Delta d$ ) of film thickness due to measuring methods defined by the following formula.

$$\Delta d = d(\text{MMV}) - d(\text{WMV})$$

[where  $d(\text{MMV})$  is the film thickness ( $\mu\text{m}$ ) determined by measuring 10 stacked films by a micrometer, and  $d(\text{WMV})$  is a thickness measured by weight ( $\mu\text{m}$ ).]

If  $\Delta d$  is more than 0.5  $\mu\text{m}$ , the roughnesses on the film surfaces form clearances between wound and stacked film layers of the capacitor, and the clearances may induce corona discharge at electrode ends, to lower dielectric strength and also to lower the electrostatic capacity unpreferably. If less than 0.01  $\mu\text{m}$ , the film may have insulation defects caused by electrostatic discharge when the film is unwound from the film roll since the contact area between film layers is large.

Furthermore, the maximum roughnesses ( $R_t$ ) on both sides of the biaxially oriented polypropylene film of the present invention are preferably 0.1 to 4.0  $\mu\text{m}$ , more preferably 0.3 to 3.0  $\mu\text{m}$ . If the maximum roughnesses are less than 0.1  $\mu\text{m}$ , slipperiness is insufficient, and air can only insufficiently escape to form longitudinal creases, etc., and windability and handling convenience may be lowered. On the other hand, if the  $R_t$  values are more than 4.0  $\mu\text{m}$ , the surfaces are too rough, and air remains in the clearance between stacked film layers, to deteriorate the capacitor

device, and the film may be broken, to lower productivity.

In the present invention, the crystallinity obtained from the density of the biaxially oriented polypropylene film is preferably 70% or more, more preferably 72% or more, further more preferably 74% or more. If the crystallinity is less than 70%, the biaxially oriented polypropylene film may decline in heat resistance, dielectric properties and metallizability.

The biaxially oriented polypropylene film of the present invention is preferably 1.5 to 3.5% in the sum of heat shrinkage in the machine and transverse directions when heated at 120°C for 15 minutes. If the sum of heat shrinkage is too large, dimensional change may occur to crease the film roll when a metallic layer is formed as an electrode, and the inside of the film and/or the portion in contact with an external electrode may be stressed because of too large mechanical deformation caused when the capacitor device is produced, to greatly lower the capacity of the capacitor or to destroy the device. If the sum of heat shrinkage percentages is too small, the winding tightening by the heat treatment when the capacitor device is produced is insufficient, to adversely affect the form integrity and the capacity change rate. The sum of heat shrinkage is more preferably 1.6 to 3.3%, further more preferably 1.7 to 3.0%, still further more preferably 1.8 to 2.8%, especially preferably 1.8 to 2.5%.

Especially when the metallized layer is changed in thickness in the transverse direction of the film as described later, to increase the thickness of the portion in contact with an external electrode, the film roll has portions different in metallized layer thickness formed in stripes in the machine direction of the film. So, if the heat shrinkage is large, the film roll is greatly creased, and in the processing of a capacitor device, the slit edge face may not be aligned, to make the connection with the external electrode insufficient, lowering electric current resistance as a problem in actual use.

In the present invention, the thickness of the biaxially oriented polypropylene film (the film thickness determined by measuring 10 stacked films using a micrometer) is preferably 2.0 to 30  $\mu\text{m}$ , more preferably 2.5 to 20  $\mu\text{m}$  in view of film formability, mechanical properties and electrical properties. If the film thickness is too small, the dielectric strength and mechanical strength may become insufficient, and the film may be damaged by metallization, especially by heat. If the thickness of the film is too large, it is difficult to form a film with a uniform thickness, and when the film is used as the dielectric of a capacitor, the capacity per unit volume becomes small unpreferably.

The thickness of the film is selected to suit the application concerned, considering whether the power source is DC or AC, the working voltage, and the size and capacity of the capacitor. For a capacitor used in AC, especially for an AC circuit required to be high in heat resistance, the film thickness is preferably 3 to 10  $\mu\text{m}$ . On the other hand, for a capacitor used in DC, especially for a high frequency circuit used with a large current, that is, for an application where electric current resistance is required, the thickness is preferably 3 to 10  $\mu\text{m}$ , and especially when high dielectric strength is required, the thickness is preferably 5 to 15  $\mu\text{m}$ . For a DC circuit required to be high in heat resistance, the thickness is preferably 3 to 15  $\mu\text{m}$ , and for electric power uses, the thickness is preferably 15 to 25  $\mu\text{m}$ .

The biaxially oriented polypropylene film of the present invention is preferably 5 to 12% in the weight change rate when immersed in dodecylbenzene. The weight change rate corresponds to the swellability of the film. If the weight change rate exceeds 12%, the film is swollen greatly at the device ends through which an insulating oil is impregnated when the capacitor device is impregnated with an insulating oil, to choke the moderate clearance between the film layers where the insulating oil is going to permeate through. So, it is prevented that the insulating oil is impregnated deep inside the device, not allowing the effect of the insulating oil to give corona resistance to be sufficiently manifested, and the swelling may excessively stress the film used as the dielectric of the capacitor device, to lower the dielectric strength. If the weight change rate is less than 5%, since the affinity between the film surface and the insulating oil declines, the rate of the insulating oil impregnation into the clearance between the film layers declines, making it unlikely that the insulating oil uniformly fills the device internally, the effect of the insulating oil to give corona resistance may not be able to be manifested insufficiently similarly. The weight change rate is more preferably 6 to 11%, most preferably 7 to 11%.

The biaxially oriented polypropylene film of the present invention is preferably -1 to 1% in the dimension change rate in the machine direction of the film when immersed in dodecylbenzene. If the dimension change rate in the machine direction of the film exceeds 1%, winding tightening occurs when the capacitor device is impregnated with an insulating oil, to choke the moderate clearance formed between film layers, to inhibit that the insulating oil is impregnated deep inside the device, and the effect of the insulating oil to give corona resistance may not be able to be manifested sufficiently. If the dimension change rate in the machine direction of the film is less than -1% (e.g., -2%, -3%, etc.), the film is partially deflected inside the capacitor device, and the dielectric loss may be worsened. The dimension change rate in the machine direction of the film is more preferably -0.8 to 0.8%, most preferably -0.6 to 0.6%.

The biaxially oriented polypropylene film of the present invention is preferably -2 to 2% in the dimension change rate in the transverse direction of the film when immersed in dodecylbenzene. If the dimension change rate in the transverse direction of the film exceeds 2%, the contact with a sprayed metal may fail, to worsen the dielectric loss. If the dimension change rate in the transverse direction of the film is less than -2% (i.e., -3%, -4%, etc.), the film may be partially deflected inside the capacitor device, also to worsen the dielectric loss. The dimension change rate in the transverse direction of the film is more preferably -1.5 to 1.5%, most preferably -1 to 1%.

Furthermore, the biaxially oriented polypropylene film of the present invention is preferably 20 minutes or more in the oxygen absorption induction time, since the long-term durability of the capacitor using the polypropylene film as its dielectric is good.

Moreover, the biaxially oriented polypropylene film of the present invention is  $1.5 \times 10^5 \Omega\text{F}$  or more in the insulation resistance IR1 at 20°C, and 700 or less in IR1/IR2 where IR2 is the insulation resistance at 100°C, considering the case of using it as the dielectric of a capacitor. This is especially suitably used for a capacitor for a heat resistant DC circuit. It is more preferable that IR1 is  $2 \times 10^5 \Omega$  or more, and that IR1/IR2 is 500 or less. If IR1 is less than  $1.5 \times 10^5 \Omega\text{F}$ , or if IR1/IR2 exceeds 700, the dielectric properties may decline when the capacitor is used for a long time.

If a metallic layer is formed on the biaxially oriented polypropylene film of the present invention, the surface on which the metallic layer is formed is preferably treated by corona discharge or plasma for enhancing the adhesive strength. The corona discharge treatment can be carried out using any publicly known method, but the atmospheric gas of the treatment is preferably air, carbon dioxide gas, nitrogen gas or any of their mixtures. The plasma treatment can be carried out by keeping any of various gases in a plasma state for chemical modification of the film surface, and for example, the method described in Japanese Patent Laid-Open (Kokai) No. 59-98140 can be used.

The biaxially oriented polypropylene film used as the dielectric of a capacitor in the present invention can be wound together with a metallic foil used as an electrode, or can be metallized beforehand to have an electrode. For downsizing the capacitor device, a polypropylene film metallized and wound is more preferable.

When a metallic layer is formed on the biaxially oriented polypropylene film of the present invention, the metal is not especially limited, but the use of one or more in combination of aluminum, zinc, copper, tin, silver, nickel, etc. is preferable in view of the durability of the metallic layer and productivity. If a metal mainly composed of aluminum or zinc is used, an additional effect can be obtained. In the case of aluminum, the self heal properties of the capacitor at the time of dielectric breakdown is improved preferably as an additional effect, and in the case of zinc, the capacitor can have corona resistance preferably. In this case, the metal as a main ingredient means that it accounts for 50 wt% in the metals forming the metallic layer. As a preferable method for determining the contents of the individual metals actually constituting the metallic layer, at first, a certain amount of a metallic layer is dissolved in hydrochloric acid, and the contents of individual metals are determined by a plasma emission spectrometer (ICP).

If a metallic layer is formed on the biaxially oriented polypropylene film of the present invention to be used for a capacitor for AC, the metallic layer is preferably mainly composed of zinc in view of preference for corona resistance of the capacitor. If the metallic layer mainly composed of zinc is an alloy using aluminum as a metal other than zinc, the corona resistance obtained is more preferably compatible with the moderate self heal properties of the capacitor at the time of dielectric breakdown. When an alloy of zinc and aluminum is used, it is most preferable that the zinc content is 80 wt% to 95 wt% while the aluminum content is 5 wt% to 20 wt%.

On the other hand, if a metallic layer is formed on the biaxially oriented polypropylene film of the present invention to be used for a capacitor for DC, the metallic layer is preferably mainly composed of aluminum, in view of the preference for improving the self heal properties of the capacitor at the time of dielectric breakdown.

The method for forming a metallic layer on the biaxially oriented polypropylene film of the present invention can be vacuum metallization, sputtering or ion beam method, etc. but is not especially limited.

In the present invention, the metallic layer resistance of the metallized film is preferably  $1 \Omega/\text{square}$  to  $40 \Omega/\text{square}$ , more preferably  $1.2 \Omega/\text{square}$  to  $30 \Omega/\text{square}$ . If the resistance is too small, the thickness of the metallized layer is thick, and at the time of metallization, heat damage is caused to form surface defects like crater. Furthermore, if the film is as thin as about  $4 \mu\text{m}$ , holes, etc. may be formed. If the resistance is too large, the dielectric loss is worsened, and the heat generated from inside the capacitor with AC applied may not allow the long-term voltage withstand to be maintained. The layer resistance can be kept in this range preferably by monitoring the resistance of the metallic layer during metallization, to control it.

Furthermore, in the present invention, when the capacitor obtained by using the biaxially oriented polypropylene film as its dielectric is used for a heat resistant AC circuit or for a high frequency circuit in a DC circuit or for any application requiring heat resistance, the resistance value is preferably  $2 \Omega/\text{square}$  to  $10 \Omega/\text{square}$  for the reasons as mentioned above.

Moreover, in the present invention, when the capacitor obtained by using the biaxially oriented polypropylene film as its dielectric is used especially for a high frequency circuit in a DC circuit, it is preferable that the resistance of the metallic layer changes continuously in the transverse direction of the film, that is, lowest in the portion in contact with an external electrode and highest near the insulation groove, i.e., the portion to be substantially an internal electrode beyond the film dielectric. Specifically, it is preferable to form a metallic layer continuously changing in resistance in a range from minimum  $2 \Omega/\text{square}$  to maximum  $10 \Omega/\text{square}$  in the transverse direction of the film. This constitution can be achieved, for example, by placing a comb-shaped deposition preventive plate extending in the running direction of the base film between the base film and the metallization source at the time of metallization, to change the intensity of the flux of the deposited metal in the transverse direction of the film.

The film contains potential insulation defects introduced from the raw material and in the step of film formation. So,



in the production of the capacitor, a step called clearing to inactivate the insulation defects by voltage treatment is generally taken. As for the mechanism of clearing, the Joule heat by the dielectric breakdown at an insulation defect evaporates the metal electrode around it, to electrically isolate the discharge breakdown portion from the other portion of the electrode. Since the Joule heat by dielectric breakdown is constant irrespective of the deposited metal and metallic layer thickness, the likelihood for the metal electrode to evaporate affects the effect of clearing (self heal properties), and especially when the thickness of the electrode layer is smaller, the metal electrode of the nearby portions of insulation defects is perfectly evaporated to allow perfect clearing preferably.

If the clearing is imperfect, dielectric breakdown may be caused especially when a high voltage is applied. Especially when the capacitor is used at a high voltage in a high frequency circuit, the thickness of the electrode layer must be kept small for the above reason, but as a whole, if the thickness is kept small, the portion in contact with an external electrode must be inevitably small in layer thickness, to make the contact with the external electrode insufficient. As a result, the dielectric loss of the capacitor may become large. To avoid it, it is preferable that the internal electrode portion is small in layer thickness and the portions in contact with the external electrode is large in layer thickness.

In the present invention, if a metallic layer is formed on either side of the biaxially oriented polypropylene film when there is a difference between both the sides of the film in center line average surface roughness, it is preferable to form the metallic layer on the side smaller in surface roughness. In this case, the surface defects of the metallic layer can be minimized, to improve the dielectric strength and the device life.

In the present invention, the insulation groove (a portion free from the metallic layer provided on the side where the metallic layer is formed, for the purpose of electric insulation, etc.) provided when the metallic layer is formed on the biaxially oriented polypropylene film can be of ordinary type, but can also be of any of various types, for example, with a fuse mechanism to suit each object. However, especially it is preferable that the metallic layer to be an internal electrode has an insulation groove formed continuously in the machine direction of the film at one edge in the transverse direction of the film, and that the internal electrode is provided with such a safety function that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused, since the capacity change after continuous use of the capacitor can be minimized.

It is also preferable that the metallic layer as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and that such a safety function that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused is provided in such a manner that the metallic layer is separated into a plurality of isles in the machine direction of the film by insulation grooves and is connected through bottle necks with a metallic layer continuous at the other edge in the transverse direction of the film, since the dielectric loss is less worsened, and since the capacity change after continuous use of the capacitor can be minimized. This constitution can be especially preferably used in a capacitor for a heat resistant AC circuit.

Moreover, it is preferable that the metallic layer as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and that the internal electrode is provided with a plurality of such safety functions in the transverse direction of the film that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused, since the capacity change after continuous use of the capacity can be most efficiently inhibited though the layer resistance of the internal electrode becomes large to worsen the dielectric loss. This constitution can be especially preferably used in a capacitor for a heat resistant DC circuit.

The method for forming the insulation groove can be, though not limited, the laser margin method, oil margin method, etc., and the simple oil margin method can be preferably used.

In the present invention, the capacitor with metallic layers formed on biaxially oriented polypropylene films can be of the type in which two films with a metallic layer (with a margin) on one side respectively are used as a pair and wound together, and in which the pair of metallic layers are connected to two external lead electrodes without causing short-circuiting, or of the type in which a film with metallic layers (with a margin each) on both sides is wound together with one or more films not provided with any metallic layer, and in which the pair of metallic layers are connected with two lead electrodes without causing shortcircuiting. However, the type of the capacitor is not limited to them. In this case, it is preferable to connect the metallic layers provided on the films with the external lead electrodes by metal spraying.

Furthermore, in the present invention, the capacitor with metallic layers having safety functions with bottle necks formed on biaxially oriented polypropylene films is not especially limited in type, but if two metallic layers as a pair have bottle necks respectively, irrespective of whether the two metallic layers are provided on either of each film or on both sides of a film, the capacity change after continuous use of the capacitor can be kept minimum.

The types which can be adopted for the capacitor of the present invention include dry type, oil impregnated type, etc., and can be selectively adopted for respective purposes. For a capacitor for an AC circuit, oil impregnated type can



be preferably used.

The insulating oil used for an oil impregnated type capacitor is not especially limited, and can be one or more of vegetable oils, mineral oils, waxes, etc. As required, a viscosity improver, etc. can also be used together.

The film capacitor using the biaxially oriented polypropylene film of the present invention as its dielectric is preferably 200 V/ $\mu$ m or more per unit thickness in AC dielectric strength at 105°C. The reason is that while the rated AC voltage of a polypropylene film capacitor is usually 45 to 50 V/ $\mu$ m, a value of more than 4 times the voltage is preferable, considering safety. The AC dielectric strength is more preferably 210 V/ $\mu$ m or more. To keep the dielectric strength of a film capacitor in this range, it is effective to avoid creases and flaws in the processing into the capacitor.

The film capacitor using the biaxially oriented polypropylene film of the present invention as its dielectric is preferably 350 V/ $\mu$ m or more per unit thickness in DC dielectric strength at 25°C (room temperature). The reason is that while the rated DC voltage of a polypropylene film capacitor is usually 65 to 75 V/ $\mu$ m, a value of more than 4 times the voltage is preferable, considering safety. The DC dielectric strength is more preferably 370 V/ $\mu$ m or more. To keep the dielectric strength of a film capacitor in this range, it is effective to avoid creases and flaws in the processing into the capacitor.

The life of the film capacitor using the biaxially oriented polypropylene film of the present invention as its dielectric with an AC voltage of 60 V/ $\mu$ m per unit thickness (1.2 to 1.3 times the rated voltage) applied at 105°C is preferably 500 hours or more, more preferably 1000 hours or more in view of the guarantee period of the apparatus mounted with the capacitor. To keep the life in this range, it is effective to add a proper amount of an antioxidant, to heat-treat at about 100°C in the processing into the capacitor, to avoid creases and flaws in the processing into the capacitor, and to mold the capacitor in an epoxy resin or to seal the capacitor in a metallic can after impregnating it with an oil (outpackage), for intercepting the contact with the open air.

Methods for producing the biaxially oriented polypropylene film of the present invention and the capacitor using the film as its dielectric are described below, but the present invention is not limited thereto or thereby.

Raw polypropylene is supplied into an extruder, heated to be molten, filtered by a filter, and melt-extruded from a slit die at a temperature of 220 to 320°C into a film, and the film is wound on a casting drum kept at a temperature of 50 to 85°C, to be cooled and solidified, for making a cast film. In this case, if the temperature of the casting drum is too high, the film may be crystallized so intensively as to make the subsequent drawing difficult or to make the surface roughnesses too large. If lower than 50°C, the surface roughnesses may be too small. The methods which can be used for letting the film adhere to the cast drum include electrostatic application, using the surface tension of water, air knife method, press roll method, submerged casting, etc., but to obtain the polypropylene film of the present invention, the air knife method is effective since it is good in flatness and allows the control of surface roughnesses. Especially the surface temperature of the film on the cast drum side can be different from that on the opposite side due to the difference between the drum surface and air in cooling efficiency, and since the polypropylene high in stereoregularity used for the film of the present invention is high in crystallinity, the difference in surface temperature can result in the difference in the crystallinity on the film surface between both sides, to greatly affect the flatness and the difference in surface roughness between both sides. So, when the air knife method is used, it is also important to control the temperature of the blown air. To obtain the film of the present invention, it is preferable to set the temperature of the air used for the air knife method at ((Cast drum temperature) - 70°C) ~ ((Cast drum temperature) - 20°C).

Subsequently, the cast film is biaxially drawn, to be biaxially oriented. At first, the cast film is pre-heated on rolls kept at 120 to 150°C and in succession, the film is fed between pairs of rolls kept at a temperature of 140°C to 150°C and different in peripheral speed, to be drawn to 2 to 6 times in the machine direction, and immediately cooled to room temperature. If the polypropylene film of 99% or more in mmmm of the present invention is preheated at a temperature of 130°C or lower and drawn at a temperature of 140°C or lower, it may be drawn irregularly and broken, not allowing film formation due to insufficient calorific value, and it is important to adopt a drawing temperature of higher than 140°C.

In succession, the drawn film is introduced into a stenter, and drawn in the transverse direction to 5 to 15 times at a temperature of 155 to 165°C, and then while it is being relaxed by 2 to 20% in the transverse direction, it is thermally fixed at a temperature of 150 to 160°C, and wound.

Subsequently, the film is treated by corona discharge in air, nitrogen, carbon dioxide gas or any of the mixtures on the surface to be metallized, to improve the adhesive strength to the metal to be deposited, and wound by a winder.

The obtained film is set in a vacuum metallizer, and the film is coated with an oil to form an insulation groove suitable for the purpose concerned, using a gravure coater, etc. Then, the metal suitable for the purpose concerned is deposited to achieve a predetermined layer resistance. Furthermore, as required, metallization is carried out through a comb-shaped deposition preventive plate to continuously change the resistance value in the transverse direction of the film. The metallized film is slit, to make two metallized reels as a pair for making a capacitor device. Then, the reels are wound to form a device and the device is formed to be flat by a thermal press, being followed by metal spraying at the ends, attaching of leads, as required impregnation with an insulating oil, and packaging to make a capacitor.

The methods for measuring and evaluating the properties in the present invention are as described below.

## (1) Isotacticity (isotactic index: II)

A sample is subjected to extraction in n-heptane at a temperature of 60°C or lower for 2 hours to remove additives from the polypropylene, and dried in vacuum at 130°C for 2 hours. From it, a specimen of W (mg) in weight is taken and subjected to extraction in boiling n-heptane in a Soxhlet extractor for 12 hours. The specimen is then taken out, washed sufficiently in acetone, dried in vacuum at 130°C for 6 hours, and cooled to room temperature. The weight W' (mg) of the cooled specimen is measured, and the isotacticity is calculated from the following formula:

$$II = (W/W') \times 100 (\%)$$

## (2) Isotactic pentad fraction

A specimen is dissolved in o-dichlorobenzene, and <sup>13</sup>C-NMR is measured at a resonance frequency of 67.93 MHz using JNM-GX270 spectrometer produced by JEOL Ltd. The method proposed by T. Hayashi et al. [Polymer, 29, 138-143 (1988)] is used for the calculation to determine the assignment of spectral peaks and the pentad fraction. Assigning the 21.855 ppm peak to the mmmmm peak for the spectrum from the methyl group, each peak is assigned and its peak area is calculated to determine its percentage proportion relative to the total area of the peaks attributed to the methyl group. Detailed measuring conditions are as follows.

Solvent used for measurement: o-dichlorobenzene (90 wt%)/benzene-D6 (10 wt%)

Specimen concentration: 15 ~ 20 wt%

Measuring temperature: 120 ~ 130°C

Resonance frequency: 67.93 MHz

Pulse width: 10 μsec (45° pulse angle)

Pulse repetition time: 7.091 sec

Data points: 32 K

Number of accumulations: 8168

Measurement mode: Noise decoupling

## (3) Center line average surface roughness and maximum roughness (expressed as Ra and Rt respectively)

A tracer type surface roughness tester is used for measurement according to JIS B 0601. A high precision thin coating level difference measuring instrument (Model ET-10) produced by Kosaka Kenkyujo K.K. is used at a conical probe radius of 0.5 μmR, a load of 5 mg and a cut-off of 0.08 mm.

## (4) Measurement of Δd

The thickness of a film (d (MMV) (μm)) is measured by stacked 10 films using a micrometer according to JIS B 7502. A micrometer of 700 ± 100 gf and 0.0001 mm in minimum reading is used, and the value obtained by measuring 10 stacked films is divided by 10, to obtain d (MMV) (μm). To measure the film thickness by the gravimetric method (d (WMV) (μm)), a film is cut into a 10 cm square, and the weight W (g) is measured by an electronic force balance produced by Mettler. The intended weight is obtained from the following formula:

$$d \text{ (WMV)} = 100 \times W/p$$

[where d (WMV) is the gravimetric thickness (μm); W, the weight of the 10 cm square film; and p is the density (g/cm<sup>3</sup>) of the film. p is measured using ethanol-water density gravity tubes at 23 ± 0.5°C according to JIS K 7112 D method.]

Then, using the d (MMV) and d (WMV) obtained, Δd (μm) is calculated from the following formula:

$$\Delta d = d \text{ (MMV)} - d \text{ (WMV)}$$

## (5) Heat shrinkage

A sample measuring 260 mm and 10 mm in the machine and transverse directions respectively is taken out of a film, and marks are put at 30 mm from both the ends, to secure an original size (L0: 200 mm). A weight of 3 g is applied to the bottom end of the sample, and hung in an oven of 120°C, being heat-treated for 15 minutes. The sample is taken out, and the distance between the marks (L1) is measured, to calculate the heat shrinkage from the following formula. The sum of calculations in the machine and transverse direction is adopted as the heat shrinkage of the sample.

$$\text{Heat shrinkage} = [(L_0 - L_1)/L_0] \times 100 (\%)$$

(6) Ash content

5 Measurement is carried out according to JIS C 2330. A biaxially oriented polypropylene film of W0 in initial weight is put in a platinum crucible, at first combusted completely by a gas burner, and then treated in an electric furnace of 750 to 800°C for about 1 hour for perfect ashing, and the weight W1 of the obtained ash is measured. The ash content is obtained from the following formula:

$$10 \quad \text{Ash content} = (W_1/W_0) \times 1000000 \text{ (ppm)}$$

W0: Initial weight (g)

W1: Ash weight (g)

15 (7) Insulation defect test

A film of 7.5 μm in thickness obtained by the 10-film micrometer method according to JIS C 2330 is judged in reference to the following criterion:

- 20 o : Two or less insulation defects  
 Δ : Three to five insulation defects  
 x : Six or more insulation defects (the film cannot be used for a capacitor.)

In the present invention, a film judged to be o or Δ is acceptable.

25 (8) Dielectric strength of device (AC)

A capacitor device kept at 105°C in a hot air oven is connected to an AC high voltage stabilizing power supply (frequency 60 Hz) produced by Kasuga Denki Co., Ltd., and a voltage increased at a rate of 200 V/sec is applied, to identify the voltage at which the device has been destroyed. The average value of 10 samples is adopted as the dielectric strength of the device which is judged in reference to the following criterion:

- 30 o : 200 V/μm or more  
 Δ : 150 V/μm to less than 200 V/μm  
 35 x : Less than 150 V/μm

In the present invention, the device judged to be o and Δ is acceptable.

(9) Dielectric strength of device (DC)

40 A capacitor device kept at 25°C is connected to a DC high voltage stabilizing power supply produced by Kasuga Denki Co., Ltd., and a voltage increased at a rate of 200 v/sec is applied to identify the voltage at which the device has been destroyed. The average value of 10 samples is adopted as the dielectric strength of the device.

45 (10) Device life test

An AC voltage of 60 V/μm (frequency 60 Hz) per film thickness is applied to a capacitor device in an atmosphere of 105°C, to measure the time till the device is destroyed. The device is judged in reference to the following criterion:

- 50 o : 500 hours or more  
 Δ : 400 hours to less than 500 hours  
 x : Less than 400 hours

In the present invention, the device judged to be o or Δ is acceptable.

55 (11) Weight change rate and dimension change rate after immersion in dodecylbenzene

A film sample is treated at 100°C in dodecylbenzene without any load applied for 8 hours, and is naturally allowed

to cool at room temperature taking 16 hours, and this cycle is repeated 3 times, to obtain the respective change rates of the film from the following formulae. In this case, two dimension change rates in the machine direction (MD) and the transverse direction (TD) of the film are measured. Each sample is measured at 5 places, and the average value is adopted.

$$\text{Weight change rate (\%)} = (J_0 - J_1)/J_0 \times 100$$

$$\text{Dimensional change rate (\%)} = (M_0 - M_1)/M_0 \times 100$$

(where  $J_0$  and  $J_1$  are the weights of the film sample before and after immersion in dodecylbenzene respectively, and  $M_0$  and  $M_1$  are the dimensions of the film sample before and after immersion in dodecylbenzene.)

#### (12) Evaluation of corona resistance

An AC voltage of 30 V/ $\mu$ m (frequency 60 Hz) per film thickness is applied to a capacitor device in an atmosphere of 105°C for 500 hours, and the capacity change rate of the device is obtained from the following formula. The device is evaluated in reference to the following criterion:

$$\text{Volume change rate (\%)} = (C_1 - C_0)/C_0 \times 100$$

(where  $C_0$  is the capacity of the capacitor device before voltage application, and  $C_1$  is the capacity of the capacitor device after voltage application.)

- o : Capacity change rate < 5% Corona resistance is good.
- Δ : 5% ≤ Capacity change rate ≤ 10% Corona resistance does not pose any problem.
- x : 10% < Capacity change rate Corona resistance is insufficient.

In the present invention, the device judged to be o or Δ is acceptable.

#### (13) Electric current resistance

A high frequency constant current power supply (HFS100K-100 produced by Takasago) is used, to feed a current of 10 A to a capacitor device at 100 kHz at a voltage of 10 V for 3 minutes, and the dielectric loss of the sample is measured according to the Schering bridge method.

#### (14) Insulation resistance and self heal properties

The insulation resistance of a capacitor device is measured according to Electronic Industries Association of Japan Standard EIAJ RC-3666A (IR1 and R0). Then, DC 500 V is applied, and the voltage is increased in steps of DC 100 V, to be applied for 1 minute each, and the number of discharge breakdown points is observed in reference to discharge sounds. When 10 discharge breakdown points have been observed, the voltage application is stopped. The insulation resistance (R) after discharge breakdown is measured, and the device is judged in reference to the following criterion with the R/R0 value as the self heal properties indicator.

- R/R0 1.5 or more : Self heal properties oo
- R/R0 1 to less than 1.5 : Self heal properties o
- R/R0 0.8 to less than 1 : Self heal properties Δ
- R/R0 Less than 0.8 : Self heal properties x

In the present invention, a device judged to be oo, o or Δ is acceptable.

#### (15) Resistance of metallic layer

A metallized film sample is cut in a piece of 2 mm in width and 50 mm in length in the machine direction, and the resistance value in the machine direction is measured according to the four-point contact method, and calibrated in reference to the width and the voltage measuring terminal distance. For a sample different in resistance in the transverse direction, the sample was measured in the entire width, to obtain the maximum and minimum values.

## (16) Oxygen absorption induction time

Three grams of a polypropylene film cut into pieces is placed in an oxygen atmosphere of one atmospheric pressure, and heated at 184°C. The oxygen pressure is measured with the lapse of time, and the time till the pressure suddenly begins to decrease is adopted as the oxygen absorption induction time.

## (17) Long-term DC application durability of capacitor

A capacitor device is encased in a plastic case together with an epoxy resin used as a filler, to produce a capacitor sample. A DC voltage of 200 V/μm is applied to the capacitor sample in an atmospheric temperature of 85°C for 1000 hours, and the numbers of insulation defects before and after voltage application are counted.

The present invention is described below in detail in reference to examples of the present invention and comparative examples.

## Example 1

A mixture with 3000 ppm of 2,6-di-t-butyl-p-cresol (BHT) and 4000 ppm of tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane (Irganox 1010) added to raw polypropylene of 99.1% in II, 99.7% in mmmm, 21 ppm in ash content and 4.2 g/10 min in melt flow rate was supplied into an extruder, molten at a temperature of 280°C, and extrusion-molded into a sheet from a T die, and the sheet was wound on a casting drum of 80°C using the air knife method at an air temperature of 25°C, to be cooled and solidified. Then, the sheet was preheated at 143°C, and fed between pairs of rolls kept at 148°C and different in peripheral speed, to be drawn to 4.8 times in the machine direction. In succession, the film was introduced into a stenter and drawn to 11 times in the transverse direction at 161°C, and then, being relaxed by 10% in the transverse direction, it was heat-treated at 150°C, to obtain a 7.50 μm thick biaxially oriented polypropylene film. Furthermore, it was treated by corona discharge in air at a treatment intensity of 30 W · min/m<sup>2</sup>. The obtained film was 99.0% in II and 99.6% in mmmm. The film was set in a vacuum metallizer, and with copper as a nucleating metal, zinc containing 8wt% of aluminum was deposited on the corona treated surface to achieve a layer resistance of 4.0 Ω/square. The metallization was carried out by the oil margin method in such a manner that an insulation groove 2 might be formed (margin; 1 mm in the length in the transverse direction) at one edge in the transverse direction of the metallic layer 1 as shown in Fig. 1 after slitting.

The film was slit to obtain a metallized film of 38 mm in overall width. Two reels of the obtained film as one pair were wound to form a device, and the device was sprayed with a metal at the end faces. Leads were attached there, to produce a capacitor device of 5 μF in capacity. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

## Comparative example 1

A polypropylene film (97.8% in II and 99.1% in mmmm) and a capacitor device were obtained as described in Example 1, except that raw polypropylene of 98.0% in II, 99.2% in mmmm, 21 ppm in ash content and 3.1 g/10 min in melt flow rate was used and that the cast drum temperature was 85°C. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

## Comparative example 2

The production of a polypropylene film was attempted as described in Example 1 using raw polypropylene of 99.8% in II, 99.9% in mmmm, 12 ppm in ash content and, 2.4 g/10 min in melt flow rate, but after drawing in the transverse direction, film breaking occurred frequently, not allowing the film to be obtained stably. The film which could be produced without being broken was 99.7% in II and 99.9% in

## Comparative example 3

A polypropylene film (98.2% in II and 98.5% in mmmm) and a capacitor device were obtained as described in Example 1, using raw polypropylene of 98.4% in II, 98.9% in mmmm, 21 ppm in ash content and 3.9 g/10 min in melt flow rate. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

## Comparative example 4

A polypropylene film (99.0% in II and 99.6% in mmmm) and a capacitor device were obtained as described in

Example 1, using the same raw polypropylene as used in Example 1, except that the cast drum temperature was 90°C. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

#### Comparative example 5

A polypropylene film (99.0% and 99.6% in mmmm) and a capacitor device were obtained as described in Example 1, using the same raw polypropylene as used in Example 1, except that the cast drum temperature was 30°C. The evaluation results of the obtained polypropylene film and capacitor are shown together in Table 1.

#### Example 2

A polypropylene film (99.0% in II and 99.6% in mmmm) and a capacitor device were obtained as described in Example 1, using the same raw polypropylene as used in Example 1, except that the cast drum temperature was 70°C. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

#### Example 3

A polypropylene film (99.0% in II and 99.6% in mmmm) and a capacitor device were obtained as described in Example 1, using the same raw polypropylene as used in Example 1, except that the cast drum temperature was 80°C and that the air temperature of the air knife was 0°C. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

#### Example 4

A polypropylene film (99.0% in II and 99.6% in mmmm) and a capacitor device were obtained as described in Example 1, using the same raw polypropylene as used in Example 1, except that the cast drum temperature was 70°C and that the air temperature of the air knife was 50°C. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

#### Example 5

A polypropylene film (99.0% in II and 99.6% in mmmm) and a capacitor device were obtained as described in Example 1, using the same raw polypropylene as used in Example 1, except that the air knife was not used. During film formation, the film was curled in the transverse direction, and during transverse drawing, declipping sometimes occurred to lower the yield. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

#### Example 6

A polypropylene film (98.6% in II and 99.2% in mmmm) and a capacitor device were obtained as described in Example 1, except that the raw polypropylene used was obtained by dry-blending the raw polypropylene used in Example 1 and the raw polypropylene used in Comparative Example 1 at 1 : 2. The evaluation results of the obtained polypropylene film and capacitor device are shown together in Table 1.

Table 1

	Example 1	Comparative example 1	Comparative example 3	Comparative example 4	Comparative example 5	Example 2	Example 3	Example 4	Example 5	Example 6
Ra ( $\mu\text{m}$ ) on drum side	0.18	0.18	0.15	0.53	0.005	0.08	0.17	0.09	0.20	0.15
Ra ( $\mu\text{m}$ ) on non-drum side	0.30	0.21	0.20	0.38	0.01	0.08	0.10	0.14	0.37	0.18
d(MMV) ( $\mu\text{m}$ )	7.50	7.50	7.50	7.50	7.50	7.50	7.50	7.50	7.50	7.50
d(WMV) ( $\mu\text{m}$ )	7.22	7.34	7.43	6.89	7.50	7.38	7.09	7.15	7.02	7.27
$\Delta d$ ( $\mu\text{m}$ )	0.38	0.16	0.07	0.61	0.00	0.12	0.41	0.35	0.48	0.23
Heat shrinkage (%)	2.2	3.8	4.2	1.8	2.7	2.3	2.6	2.4	2.1	3.1
Dielectric strength of device (V/ $\mu\text{m}$ )	215	135	120	95	105	225	180	190	185	173
Life of device (hours)	680	280	270	180	160	1350	580	620	520	470
Insulation defects	$\Delta$	$\bigcirc$	$\bigcirc$	$\times$	$\times$	$\bigcirc$	$\Delta$	$\bigcirc$	$\Delta$	$\bigcirc$



## Example 7

The film used in Example 1 was set in a vacuum metallizer, and with copper as a nucleating metal, zinc (containing 8 wt% of aluminum) was deposited on the corona treated surface to achieve a layer resistance of  $4.0 \Omega/\text{square}$ . In this case, the metallization was carried out in such a manner that the metallic layer 1 was formed to be provided with an insulation groove 2 (margin: 1 mm in the length in the transverse direction) continuous in the machine direction of the film at one edge in the transverse direction of the film according to the oil margin method as shown in Fig. 2 after slitting, and that the metallic layer 1 was provided as a plurality of isles at 30 mm intervals in the machine direction of the film by insulation grooves 3 (1 mm in the width in the machine direction), and connected by a continuous metallic layer (1 mm in width) at the other edge in the transverse direction of the film and bottle necks 4 (1 mm in both machine and transverse directions). The film was slit to obtain a metallized film of 38 mm in overall width. The obtained film and the metallized film used in Example 1 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there, to produce a capacitor device of  $5 \mu\text{F}$  in capacity, and the capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

## Example 8

A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Example 2 was used. The obtained film and the metallized film used in Example 2 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there, to produce a capacitor device of  $5 \mu\text{F}$  in capacity, and the capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

## Example 9

A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Example 3 was used. The obtained film and the metallized film used in Example 3 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there, to produce a capacitor device of  $5 \mu\text{F}$  in capacity, and the obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

## Example 10

A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Example 5 was used. The obtained film and the metallized film used in Example 5 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device of  $5 \mu\text{F}$  in capacity, and the obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

## Example 11

A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Example 6 was used. The obtained film and the metallized film used in Example 6 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device of  $5 \mu\text{F}$  in capacity, and the obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

## Example 12

A capacitor was produced as described in Example 7, except that the metallized film used in Example 1 was used as two reels of a pair. The evaluation results of the obtained capacitor are shown together in Table 2.

## Comparative example 6

A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Comparative Example 1 was used. The obtained film and the metallized film used in Comparative Example 1 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were

attached there to produce a capacitor device of 5  $\mu\text{F}$  in capacity, and the obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

#### Comparative example 7

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A metallized film was produced as described in Example 7, except that the biaxially oriented polypropylene film of Comparative Example 4 was used. The obtained film and the metallized film used in Comparative Example 4 were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device of 5  $\mu\text{F}$  in capacity, and the obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

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#### Example 13

A capacitor device was produced as described in Example 7, except that aluminum was deposited as an internal electrode to achieve a layer resistance of 4.0  $\Omega/\text{square}$ . The obtained capacitor device was impregnated with crystalline wax, to obtain a capacitor. The evaluation results of the capacitor are shown together in Table 2.

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Table 2

	Example 7	Example 8	Example 9	Example 10	Example 11	Example 12	Example 13	Comparative example 6	Comparative example 7
Weight change rate (%)	11.5	10.5	10.8	9.7	14.1	10.5	10.5	13.5	10.5
Dimension change rate (MD, %)	0.2	0.3	0.7	1.2	-0.1	0.3	0.3	1.5	1.6
Dimension change rate (TD, %)	-0.1	-0.2	-3.2	0.3	-1.8	-0.2	-0.2	0.2	0.5
Corona resistance	O	O	O	Δ	Δ	Δ	Δ	Δ	O
Self heal properties	O	O	O	O	Δ	Δ	O	Δ	O
Dielectric strength of device (V/μm)	220	240	230	203	180	230	250	140	180
	O	O	O	O	Δ	O	O	x	Δ
Life of device (hours)	1050	1800	950	880	730	700	490	280	200
	O	O	O	O	O	O	Δ	x	x
Insulation defects	Δ	O	Δ	O	Δ	O	O	O	x

## Example 14

Polypropylene of 98.8% in II, 99.4% in mmmm and 19 ppm in ash content was supplied to an extruder, molten at a resin temperature of 280°C, and extrusion-molded as a sheet from a T die, and the sheet was wound around a casting drum of 70°C using the air knife method at an air temperature of 25°C, to be cooled and solidified, preheated to 135°C, and fed between pairs of rolls kept at 140°C and different in peripheral speed, to be drawn to 5 times in the machine direction, and immediately cooled to room temperature. In succession, the sheet was introduced into a stenter, preheated to 170°C, drawn to 10 times in the transverse direction at 165°C, and being relaxed by 8% in the transverse direction, treated by corona discharge in air at 30 W · min/m<sup>2</sup>. The ash content and mmmm of the obtained film were not different from those of the raw polypropylene. The film was set in a vacuum metallizer, and with copper as a nucleating metal, zinc was deposited on the corona treated surface to achieve a layer resistance of 4.0 Ω/square, in such a manner that a metallic layer 1 was formed with an insulation groove 2 (margin: 1 mm in the length in the transverse direction) provided at one edge in the transverse direction. The film was slit, to obtain a metallized film of 38 mm in over-all width.

The obtained film was used as two reels of a pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device of 5 μF in capacity. The properties of the obtained film and the capacitor device are shown in Table 3. The film was excellent in dielectric properties. The capacitor device was also good in dielectric properties and dielectric loss after application of large current, being judged to be Δ in self heal properties.

Examples 15 to 17, and comparative examples 8 to 11

As shown in Table 3, biaxially oriented polypropylene films and capacitors different in respective conditions were obtained as described in Example 14.

The properties of the films and capacitors are shown in Table 3.

In the table, the respective abbreviations mean the following:

BDV: Breakdown voltage

DC: Direct current

Table 3

	I I (%)	monum (%)	Ash content (ppm)	Ra ( $\mu$ m) on drum side/no n-drum side	d(MMV) ( $\mu$ m)	Crystallinity (%)	BDV (V/ $\mu$ m)	Heat shrinkage (%)	BDV of device DC(V/ $\mu$ m)	tan $\delta$ after application of large current	Self heal properties
Example 14	98.8	99.5	19	0.08/0.08	5.0	76	640	1.9	380	0.06	$\Delta$
Example 15	98.9	99.2	19	0.08/0.08	5.0	75	620	2.3	355	0.06	$\Delta$
Example 16	98.7	99.0	21	0.08/0.08	5.0	71	615	2.4	345	0.07	$\Delta$
Example 17	98.8	99.5	19	0.08/0.08	3.5	74	595	2.2	340	0.07	$\Delta$
Comparative example 8	98.0	98.5	25	0.08/0.08	5.0	70	550	2.6	305	0.06	$\Delta$
Comparative example 9	98.2	98.4	20	0.08/0.08	3.5	67	450	3.0	280	0.07	$\Delta$
Comparative example 10	97.5	98.8	19	0.08/0.08	5.0	66	600	2.8	310	0.07	$\Delta$
Comparative example 11	98.2	99.2	38	0.08/0.08	5.0	72	460	2.0	280	0.11	$\Delta$

Examples 15 to 17 conform to the present invention. The obtained biaxially oriented polypropylene films were good in dielectric properties, and the capacitors obtained using the films were excellent in dielectric properties, good in die-

lectric loss after application of large current, and judged to be  $\Delta$  in self heal properties.

On the other hand, Comparative Examples 8 to 10 do not conform to the present invention in mmmm. The obtained biaxially oriented polypropylene films were small in dielectric breakdown voltage, and the capacitors obtained using the films were poor in dielectric properties, and furthermore large in heat shrinkage, being inferior also in heat resistance.

Comparative Example 11 does not conform to the present invention in ash content though it conforms to the present invention in mmmm. It was poor in dielectric properties and dielectric loss after application of large current.

Examples 18 to 20

The same biaxially oriented polypropylene film as used in Example 14 was used, and a comb-shaped deposition preventive plate was inserted between the metallization source and the base film at the time of metallization, to let the layer resistance change continuously in the transverse direction of the film from 2.5  $\Omega$ /square to 8  $\Omega$ /square. A margin was formed on the electrode metal side of 8  $\Omega$ /square. Two pairs of such symmetrical films were processed into a capacitor device as described in Example 14 (Example 18).

In Example 19, the layer resistance was kept constant at 12  $\Omega$ /square in the transverse direction of the film. In Example 20, the layer resistance was kept constant at 1.5  $\Omega$ /square in the transverse direction of the film. The results are shown in Table 4.

Example 21

A capacitor device was produced as described in Example 18, except that the metal used for metallization was aluminum. The results are shown in Table 4.

Table 4

	I, I (%)	minum (%)	Ash content (ppm)	Ra ( $\mu\text{m}$ ) on drum side/no n-drum side	d (MMV) ( $\mu\text{m}$ )	Maximum layer resistance ( $\Omega/\square$ )	Minimum layer resistance ( $\Omega/\square$ )	BDV of device ( $\text{V}/\mu\text{m}$ )	tan $\delta$ after application of large current	Self heal properties
Example 18	98.8	99.5	19	0.08 / 0.08	5.0	8	2.5	380	0.02	○
Example 19	98.8	99.5	19	0.08 / 0.08	5.0	12	12	385	0.08	○
Example 20	98.8	99.5	19	0.08 / 0.08	5.0	1.5	1.5	380	0.01	x
Example 21	98.8	99.5	19	0.08 / 0.08	5.0	8	2.5	380	0.01	⊙

All the capacitor devices showed excellent dielectric breakdown voltage. Especially the capacitor device of Example 18 was excellent also in the dielectric loss after application of large current and self heal properties, being very



excellent as a capacitor for a high frequency circuit. The capacitor device of Example 19 was large in layer resistance, and rather higher in the dielectric loss after application of large current, compared to Example 18. The capacitor device of Example 20 was low in layer resistance, and rather inferior in self heal properties compared to Example 18. The capacitor device of Example 21 was better in self heal properties compared to Example 18, being most excellent as a capacitor for a high frequency circuit.

#### Example 22

The raw polypropylene used in Example 2 was used to obtain a 5.0  $\mu\text{m}$  thick biaxially oriented polypropylene film as described in Example 1. The obtained film was set in a vacuum metallizer, and aluminum was deposited on the corona treated surface, with an insulation groove 2 (margin: 2.5 mm in the length in the transverse direction) provided at one edge in the transverse direction of the metallic layer 1 as shown in Fig. 1 to achieve a layer resistance of 8  $\Omega/\text{square}$ . The metallized film was slit, to obtain a metallized film of 100 mm in overall width.

Two reels of the obtained metallized film were used as one pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device, and the obtained capacitor device was sealed in a plastic case together with an epoxy resin used as a filler. In this way, 10 capacitors were produced. The electrostatic capacity of the capacitors was 60  $\mu\text{F}$ .

The capacitors were impressed at a DC voltage of 1000 V in an atmospheric temperature of 85°C for 100 hours, and the number of capacitors which suffered any dielectric breakdown was counted.

The capacitors which did not suffer any dielectric breakdown in the above test were unwound to obtain metallized polypropylene films, and the number of insulation defects of each metallized polypropylene film (IF) was counted, while the number of insulation defects of a metallized polypropylene film not energized (IF0) was also counted, for examining the increase of insulation defects after impression. To count the insulation defects, a metallized polypropylene film as a specimen was held between a metallized film used as an electrode for testing and a copper flat sheet, and a DC voltage of 1.25 kV was applied for 1 minute, to count the number of breakdowns (number of defects). The area of a specimen was 0.2  $\text{m}^2$ .

The properties of the obtained film (the ash content and mmmm of the obtained film were not different from those of the raw polypropylene) and capacitor are shown in Table 5.

#### Example 23

A capacitor was produced as described in Example 22, except that the raw polypropylene used in Example 2 contained 1000 ppm of tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane as an antioxidant.

The properties of the obtained film (the ash content and mmmm of the obtained film were not different from those of the raw polypropylene) and capacitor are shown in Table 5.

#### Example 24

A capacitor was produced as described in Example 22, except that the raw polypropylene used in Example 2 contained 3000 ppm of tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane as an antioxidant.

The properties of the obtained film (the ash content and mmmm of the obtained film were not different from those of the raw polypropylene) and capacitor are shown in Table 5.

#### Example 25

A capacitor was produced as described in Example 22, except that 3000 ppm of 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene was added instead of tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane.

The properties of the obtained film (the ash content and mmmm of the obtained film were not different from those of the raw polypropylene) and capacitor are shown in Table 5.

#### Example 26

The biaxially oriented polypropylene film used in Example 25 was set in a vacuum metallizer, and aluminum was deposited on the corona treated surface to achieve a layer resistance of 8  $\Omega/\text{square}$ . In this case, a metallic layer 1 was formed to have an insulation groove 2 (margin: 2.5 mm in the length in the transverse direction) continuous in the machine direction of the film at one edge in the transverse direction of the film by the oil margin method as shown in Fig. 3 after slitting, and the metallic layer 1 had a plurality of 1 mm wide bottle necks 4 with safety function formed in the

transverse direction of the film by intermittent 20 mm long insulation grooves 3 (1 mm in width) formed at an inclination angle of 45° in the transverse direction.

Two reels of the obtained metallized film were used as a pair and wound to form a device, and the end faces of the device were sprayed with a metal. Leads were attached there to produce a capacitor device, and the obtained capacitor device was sealed in a plastic case together with an epoxy resin used as a filler. In this way, 10 capacitors were produced. The electrostatic capacity of the capacitors was 60  $\mu$ F.

The properties of the obtained metallized film and capacitors are shown in Table 5.

#### Comparative example 12

Capacitors were produced as described in Example 22, except that the raw polypropylene of Comparative Example 3 was used.

The properties of the obtained film (the ash content and mmmm of the obtained film were not different from those of the raw polypropylene) and capacitors are shown in Table 5.

Table 5

	I I (%)	minum (%)	Ra ( $\mu\text{m}$ ) on drum side/no n-drum side	d (MMV) ( $\mu\text{m}$ )	Oxygen absorption induction time (min)	IR <sub>0</sub> ( $\Omega\text{F}$ )	IR <sub>1</sub> ( $\Omega\text{F}$ )	IR <sub>1</sub> /IR <sub>2</sub>	Number of dielectric breakdowns (per capacitor)	IF <sub>0</sub>	IF
Example 22	99.1	99.7	0.08 / 0.08	5.0	120	280000	230	1217	2	3	4
Example 23	99.1	99.7	0.10 / 0.10	5.0	15	350000	5800	60	2	2	8
Example 24	99.1	99.7	0.09 / 0.09	5.0	80	320000	900	355	1	0	2
Example 25	99.1	99.7	0.08 / 0.08	5.0	40	260000	7500	35	1	0	0
Example 26	99.1	99.7	0.08 / 0.08	5.0	40	260000	7500	35	0	0	0
Comparative example 12	98.4	98.9	0.15 / 0.20	5.0	70	120000	80	1500	9	10	Because of too many defects, they could not be counted.

## Industrial Applicability

The present invention can provide a biaxially oriented polypropylene film excellent in heat resistance and dielectric properties, less in insulation defects, and excellent in the impregnation of insulating oil into the clearance between film layers and swelling resistance when immersed in the insulating oil, and also provide a capacitor excellent in heat resistance, dielectric properties, corona resistance, long-term heat durability and electric current resistance using the polypropylene film as dielectric.

## Claims

1. A biaxially oriented polypropylene film, characterized by being 98 to 99.5% in isotacticity, 99% or more in isotactic pentad fraction, 30 ppm or less in ash content and 0.01 to 0.4  $\mu\text{m}$  in the center line average surface roughnesses on both sides.

2. A polypropylene film, according to claim 1, wherein the difference ( $\Delta d$ ) by the film thickness measuring methods defined by the following formula is 0.01 to 0.5  $\mu\text{m}$ :

$$\Delta d = d(\text{MMV}) - d(\text{WMV})$$

[where d (MMV) is the thickness of the film ( $\mu\text{m}$ ) obtained by measuring 10 films using a micrometer, and d (WMV) is the thickness measured by weight ( $\mu\text{m}$ ).]

3. A capacitor, comprising the use of the polypropylene film stated in claim 1 or 2 as dielectric.

4. A capacitor, comprising the use of the polypropylene film stated in claim 1 or 2 as dielectric, wherein a metallic layer is formed as an internal electrode at least on one side of the film.

5. A capacitor, comprising the use of the polypropylene film stated in claim 4 as dielectric, wherein the resistance value of the metallic layer is kept at 2  $\Omega/\text{square}$  to 10  $\Omega/\text{square}$ .

6. A capacitor, comprising the use of the polypropylene film stated in claim 4 or 5, wherein the main metal constituting the metallic layer is aluminum or zinc.

7. A capacitor for a heat resistant AC circuit, according to any one of claims 4 through 6, wherein the clearance between, film layers are impregnated with an insulating oil.

8. A capacitor for a heat resistant AC circuit, according to claim 7, wherein a polypropylene film of 5 to 12% in weight change rate when immersed in dodecylbenzene is used as dielectric.

9. A capacitor for a heat resistant AC circuit stated in claim 7 or 8, wherein the main metal constituting the metallic layer as an internal electrode is zinc.

10. A capacitor for a heat resistant AC circuit stated in any one of claims 7 through 9, wherein the metallic layer as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and the internal electrode is provided with such a safety function that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused.

11. A capacitor for a heat resistant AC circuit, comprising the use of the polypropylene film stated in claim 10 as dielectric, wherein the metallic layer as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and such a safety function that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused is provided in such a manner that the metallic layer is separated into a plurality of isles in the machine direction of the film by insulation grooves and is connected through bottle necks with a metallic layer continuous at the other edge in the transverse direction of the film.

12. A capacitor for a high frequency circuit, comprising the use of the polypropylene film stated in any one of claims 4 through 6, as dielectric, wherein the dielectric loss after impression with 100 kHz and 10 A for 3 minutes is 0.05% or less.

13. A capacitor for a high frequency circuit, according to claim 12, wherein the resistance of the metallic layer continuously changes in the transverse direction of the film in a range from minimum 2  $\Omega$ /square to maximum 10  $\Omega$ /square.

14. A polypropylene film, according to claim 1 or 2, which is 20 minutes or more in oxygen absorption induction time,  $1.5 \times 10^5$   $\Omega$ F or more in insulation resistance IR1 at 20°C and 700 or less in IR1/IR2 where IR2 is the insulation resistance at 100°C.

15. A polypropylene film, according to claim 14, wherein 1,3,5-trimethyl-2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)benzene and/or tetrakis[methylene-3(3,5-di-t-butyl-4-hydroxyphenyl)propionate]methane is contained by 0.05 wt% to 0.35 wt% as an antioxidant.

16. A capacitor for a heat resistant DC circuit, comprising the use of the polypropylene film stated in claim 14 or 15 as dielectric, wherein a metallic layer is provided as an internal electrode at least on one side of the film.

17. A capacitor for a heat resistant DC circuit, comprising the use of the polypropylene film stated in claim 16 as dielectric, wherein the resistance of the metallic layer is 2  $\Omega$ /square to 10  $\Omega$ /square.

18. A capacitor for a heat resistant DC circuit, comprising the use of the polypropylene film stated in claim 16 or 17 as dielectric, wherein the main metal constituting the metallic layer acting as an internal electrode is aluminum or zinc.

19. A capacitor for a heat resistant DC circuit, comprising the use of the polypropylene film stated in any one of claims 16 through 18 as dielectric, wherein the metallic layer acting as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and the internal electrode is provided with such a safety function that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused.

20. A capacitor for a heat resistant DC circuit, comprising the use of the polypropylene stated in claim 19 as dielectric, wherein the metallic layer acting as an internal electrode is formed with an insulation groove provided continuously in the machine direction of the film at one edge in the transverse direction of the film, and the internal electrode is provided with a plurality of such safety functions in the transverse direction of the film that the current flowing when the film suffers a dielectric breakdown electrically insulates the internal electrode portion around the discharge breakdown point from the other internal electrode portion free from any discharge caused.

FIG. 1

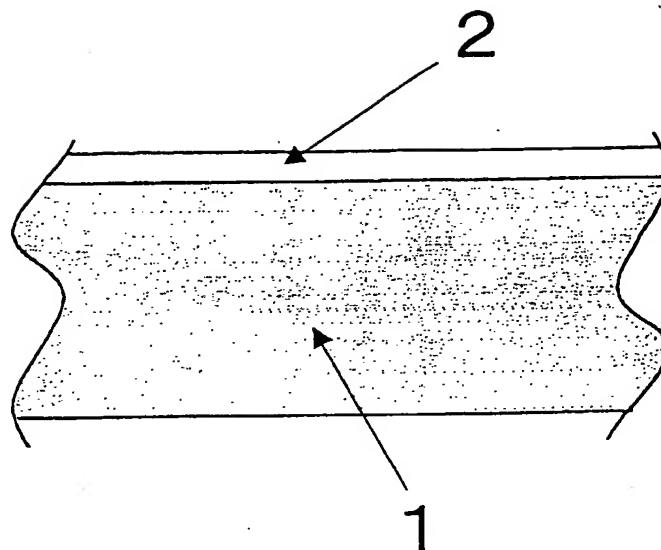


FIG. 2

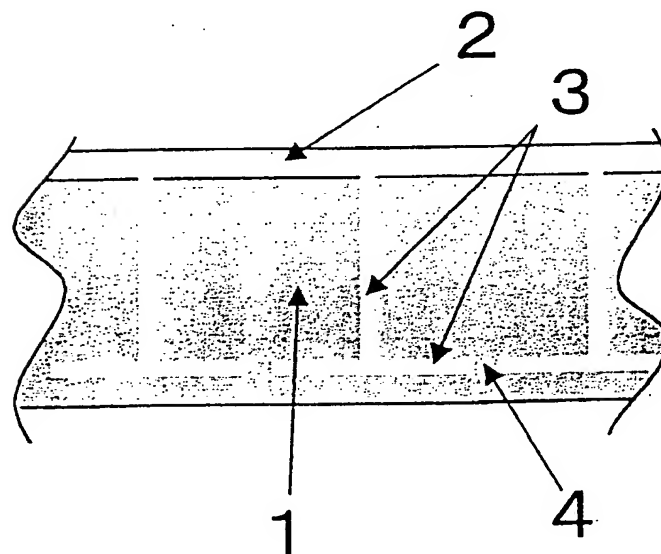
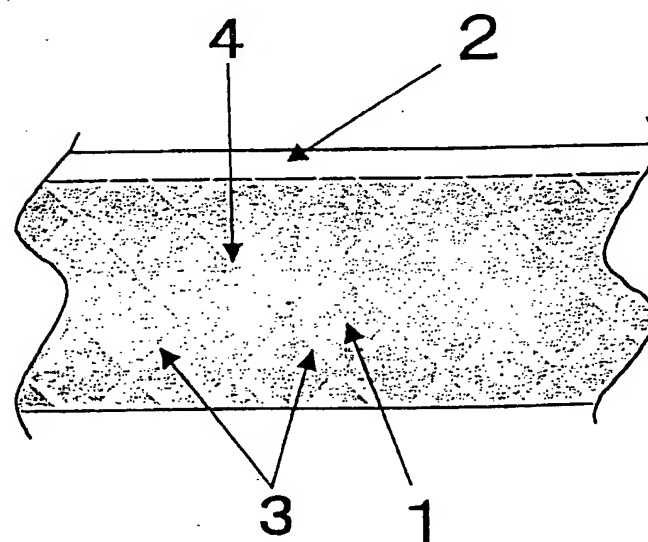


FIG. 3



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP97/02791

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> Int. Cl <sup>6</sup> C08J5/18, B29C55/12, H01G4/18 // B29K23:00, B29L7:00, C08L23:00 According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) Int. Cl <sup>6</sup> C08J5/18, B29C55/12, H01G4/18 // B29K23:00, B29L7:00, C08L23:00 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1926 - 1997 Kokai Jitsuyo Shinan Koho 1971 - 1997 Toroku Jitsuyo Shinan Koho 1994 - 1997 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP, 7-201648, A (Shin-Etsu Chemical Co., Ltd.), August 4, 1995 (04. 08. 95), Claim (Family: none)	1 - 20
A	JP, 8-156118, A (Mitsui Toatsu Chemicals, Inc.), June 18, 1996 (18. 06. 96), Claim (Family: none)	1 - 20
A	JP, 8-157613, A (Toray Industries, Inc.), June 18, 1996 (18. 06. 96), Claim (Family: none)	1 - 20
A	JP, 4-67330, B2 (Matsushita Electric Industrial Co., Ltd.), October 28, 1992 (28. 10. 92), Claim; Fig. 3 (Family: none)	1 - 20
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search October 28, 1997 (28. 10. 97)		Date of mailing of the international search report November 11, 1997 (11. 11. 97)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.		Authorized officer  Telephone No.

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